

Design Issues for Using Magnetic Materials in Radiation Environments at Elevated Temperature

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Abstract. One of the challenges of designing motors and alternators for use in nuclear powered space missions is accounting for the effects of radiation. Terrestrial reactor power plants use distance and shielding to minimize radiation damage but space missions must economize volume and mass. Past studies have shown that sufficiently high radiation levels can affect the magnetic response of hard and soft magnetic materials. Theoretical models explaining the radiation-induced degradation have been proposed but not verified. This paper reviews the literature and explains the cumulative effects of temperature, magnetic-load, and radiation-level on the magnetic properties of component materials. Magnetic property degradation is very specific to alloy choice and processing history, since magnetic properties are very much entwined with specific chemistry and microstructural features. However, there is basic theoretical as well as supportive experimental evidence that the negative impact to magnetic properties will be minimal if the bulk temperature of the material is less than fifty percent of the Curie temperature, the radiation flux is low, and the demagnetization field is small.

Keywords: Magnets, Permanent Magnets, Power Converters, Nuclear Electric Power Generation, Radiation Tolerance.

INTRODUCTION

The United States successfully flew a space fission power system in 1965 and the Soviet Union launched several trial fission power plants. The power needs of the space science community in the ensuing years have been met primarily by solar power conversion and radioisotope power conversion. As missions journey further from the Sun, require more power, and the stockpile of ²³⁸Pu radioisotope sources are depleted, mission designers are once again considering fission power options. In the past decade, space mission designers have reviewed and proposed numerous fission power system for a range of mission scenarios. In each case there are multiple design options available and there are always some technologies that require development to extend the terrestrial fission-technology base. This paper addresses one design challenge of using motors and alternators in the higher radiation environments necessary for a compact fission power system. The existing radiation hardness data for magnetic materials can provide guidelines that can be used when developing motor/alternator designs and selecting component materials. This analysis began with scrutinizing the radiation tolerance of component materials within the linear alternator of a Stirling power convertor [1], but is extensible to other magnetic material applications.

MAGNETIC MATERIAL PROPERTIES

All materials respond to applied magnetic fields, but the degree and the usefulness of the magnetic response varies greatly. Atoms that have full electron shells and subshells have only a weak, diamagnetic response to an applied magnetic field. This negative response is due to the applied field's influence on the angular momentum of orbiting electrons [2]. Atoms without completely filled shells/subshells have a permanent magnetic moment based on incomplete cancelation of electron-spin moments and can be either paramagnetic or ferromagnetic. The atomic-level magnetic moments for paramagnetic materials are random and these materials have no macroscopic magnetic strength except in the presence of an external field. Ferromagnetic materials, however, may possess a permanent magnetic moment in the absence of an external field due to the physical alignment of electron-spin moments in adjacent atoms. These atoms with aligned spin moments are atomic dipoles and are the fundamental building block

of a magnetic domain. Observations have shown a clear trend between electron structure and the magnetic strength. The narrow d-shell bands in certain transition metals and rare earth metals are favorable for magnetism [2].

The system magnetic field vector, \mathbf{B} , is the sum of the external magnetic field, \mathbf{H} , and the material's magnetic field, \mathbf{M} . Figure 1 illustrates the magnetic material response exploited in an electric motor/generator. The material starts in a demagnetized state ($\mathbf{B} = \mathbf{H} = 0$). The application of a positive external field, \mathbf{H} , leads to an increase in measured magnetic field, \mathbf{B} . A similar plot can be drawn for the material magnetic field response, \mathbf{M} , versus external field. As the external field strength increases there is increasingly difficult realignment of magnetic domains until saturation is reached, B_s or M_s . When the applied field is reversed, the magnetic domains once again rotate along easy directions first and produce a response curve with varying degrees of hysteresis. The initial slope of the curve is the initial permeability, μ_i , and μ is the proportionality constant between B and H . The residual induction, B_r , or remanence, M_r , represent the remaining magnetic field strength when there is no applied field. The coercivity, H_c , is the reverse (or negative) external field strength that must be applied to return B to zero. Similarly the intrinsic coercive field strength, H_{ci} , is the applied field strength needed to return M to zero. The shape of this B - H curve and the values of B_s , B_r , H_c , and μ_i describe the engineering properties of the magnetic material.

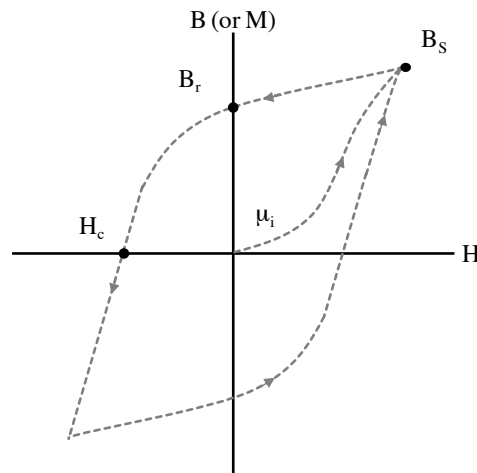


FIGURE 1. Illustration of Applied Field Strength, H , plotted versus Magnetic Flux Density, B . Soft magnetic materials should have a large permeability, μ_i , and low coercivity, H_c , to minimize the hysteresis loop and hence energy loss on each cycle. High coercivity, remanence, B_r , saturation, B_s , and low permeability are desirable for hard magnets.

At a microstructural level, as the magnet domain (region of aligned dipoles) increases, the magnetic strength of the material grows. There is a maximum possible magnetic strength of each ferromagnetic material based on its atomic structure. Therefore, chemistry changes either can raise or lower the magnetic saturation (B_s , M_s) of a given alloy depending on how the minor elemental atom(s) incorporate into the host lattice. The magnetic saturation is also a function of temperature because increasing thermal energy causes atomic vibration which opposes the magnetic alignment forces. The Curie temperature, T_c , is the temperature at which dipole spin alignment no longer occurs to a significant degree. Although the measurable magnetic strength disappears at the Curie temperature, it is interesting to note that neutron scattering experiments show that some very localized magnetic moments still exist [8].

The periodic alignment at the atomic level, also known as the crystallographic structure, influences many of the magnetic parameters. Magnetic domains are bounded by domain walls that are similar to crystallographic grain boundaries in that they move more freely as temperature rises and their motion can be limited, or pinned, by microstructural features. Limiting grain boundary size is one common way to control magnetic domain size. Magnetic permeability, susceptibility (the proportional constant between M & H) and coercivity (remnant strength at $B = 0$) are affected by microstructure as well as chemistry and temperature. The influence of the crystal structure on magnetic reorientation has to do both the metallic crystal symmetry and the atomic spin orbital shape. Domain wall movement, and therefore magnetic field reorientation, is easiest along certain crystallographic directions. For example, the easy direction, or easy axis, for body center cubic iron is along the $[100]$ -type direction, which is the cube edges. The magnetic anisotropy refers to differences in the energy required to move domains along different crystallographic directions or planes. Anisotropy in the crystal lattice is important because the slope near saturation

is dominated by the last remnants of alignment and because increasing anisotropy impedes domain wall movement. A large magnetic anisotropy means that the ease of domain movement is highly orientation dependent.

Soft Magnetic Materials

Soft magnetic materials have magnetic domains that readily align to a magnetic field with minimal wasted energy. A material is considered a soft magnet when the domain wall movement and domain magnetic orientation occur in weak applied fields, that is if coercivity, H_c , is less than 1000 Amp/m [2]. In addition to low coercivity, high initial permeability is desired to have a tight B-H hysteresis loop. Induced electrical currents also lead to energy loss in soft magnetic materials, so higher electrical resistivity is a desirable material property for soft magnets. Soft magnetic material performance is improved by aligning the crystallographic directions that allow easy domain movement with the applied field directions. Soft magnetic material performance is improved also by properties that increase remanence and lessen magnetic anisotropy. Impurities, especially in the form of precipitates, and residual stress can also pin domain walls and decrease soft magnetic performance.

Soft magnetic alloys based on equi-atomic FeCo alloys have a very high saturation strength, ~ 2.4 T, and relatively low magnetic anisotropy. This class is the preferred soft magnetic material for high flux density, high temperature applications and Hiperco 50 is a common, commercially available alloy in this class. Proper Hiperco 50 heat treatment produces the intermetallic, B2-type crystal structure with crystallographic long-range order. This intermetallic structure produces a high yield strength at elevated temperature [3, 4] but increases magnetic anisotropy and reduces ductility relative to the cubic structure without long-range order. Because this composition goes through an order-disorder phase transformation around 730 °C, the both magnetic and mechanical properties are highly sensitive to manufacturing processes. Additions of up to 2% V and 4% Ni can improve fabricability and mechanical properties with minimal negative impact on magnetic properties. For sheet products, this alloy class is typically hot rolled above 912 °C, quenched for cold rolling without long-range order, and then heat treated to regain long-range order. Rolling into thin sheets for laminations can produce crystallographic textures that impede alignment of the easy magnetization directions with applied fields. Annealing and intermediate cooling rates after annealing are important to minimize adverse crystallographic texture and optimize magnetic properties [2].

Hard Magnetic Materials

Hard magnetic materials are those which retain a relatively strong magnetic alignment and resist realignment from a reverse applied field, thus behaving as a magnetic spring pushing back on an external field. The term “permanent magnet” is sometimes used interchangeably with hard magnetic material, although no magnetic properties are permanent in a literal sense. Sufficiently high thermal energies, externally applied magnetic fields, or combinations thereof will lead to magnetic realignment and, hence, loss of “permanence”. Common permanent magnets have coercivities in the range of 10^4 to 10^6 Amp/m [2]. Magnetic permanence is enhanced by and magnetic reversal suppressed by the following:

- i) using chemistry or microstructure to increase magnetic anisotropy
- ii) maximizing remanence through preferred grain texture
- iii) pinning domain walls or making single domain particles or grains
- iv) minimizing opportunities for domain exchange between particles or grains, such as non-magnetic grain boundaries.

Magnetic coupling of transition metal elements and lanthanide series (rare earth) elements vary in a systematic and predictable manner [2]. There are several useful alloys of the RT_5 and R_2T_{17} series, where R = rare earth and T = transition metal. The phase boundary separation in the Sm-Co phase diagram is such that most SmCo alloys are in fact a combination of both $SmCo_5$ and Sm_2Co_{17} and will be generically referred to here as SmCo except when it is necessary to distinguish the phases. Based on a hexagonal structure (rather than a cubic structure), SmCo has a large magnetic anisotropy, but domain walls move readily unless pinned by some microstructural feature. The more complex, higher energy-product SmCo alloys rely on complex microstructure for grain refinement and domain pinning [5]. Alloys based on $Nd_2Fe_{14}B_1$ have a high anisotropy based on a tetragonal crystal phase, a large magnetism due to the Rare Earth-Transition Metal coupling of Nd-Fe, and the ability to grow non-magnetic B-rich

and Nd-rich phases to decouple the magnetic grains. Very small grains and preferred grain orientation are required for optimal magnetic properties. Magnetic reversal appears to be dominated by nucleation and growth of reverse domains at temperatures up to 175 °C. The Curie temperature is higher for the Co-based rare earth magnets than the Fe-based magnets. T_c also increases with increasing Co content and decreases with increasing Fe content. Apparently an antiferromagnetic exchange dominates Fe-Fe bonds at smaller bonding separation which also explains why increasing pressure can also depress T_c in Fe-based alloys [2].

MAGNETIC PROPERTIES AND RADIATION INTERACTION

When a high-energy neutron or a high-energy particle strikes a material, a large energy transfer can take place. The struck atom is referred to as the primary knock-on atom and its energy is distributed to other nearby atoms by a series of secondary collisions in what is called a displacement cascade. These collisions produce empty lattice sites (vacancies) and atoms injected into the interstices between lattice sites (interstitials). Fortunately, most vacancies and interstitials recombine; those that do not are responsible for radiation-induced microstructural and mechanical property changes. The continuous production and subsequent diffusion of point defects lead gradually to the formation of point-defect clusters and other extended defects. The evolution of such extended defects in turn is responsible for the physical and mechanical property changes of the material. Through this basic damage process, incident radiation changes defect diffusion rates, dislocation microstructures, void/bubble formation, and elemental segregation resulting in localized chemistry changes and changes in phase stability [6]. Note that radiation damage is a stochastic process and radiation can pass through a material without striking any atoms at all. Lower energy neutrons, particles, or photons are more likely to be stopped by a given target material and thus are statistically more likely to deposit their energy rather than just travel through a given material. The terms thermal and fast refer to neutron spectrums that are dominated either by lower or higher energy neutrons, respectively.

Soft Magnetic Materials and Radiation

In the early 1960s, Gordon and Sery studied the effect of radiation on numerous soft magnetic materials up to 2×10^{18} n/cm² (fast fluence $\sim 10^{17}$ n/cm²) and sample temperatures of ~ 80 °C [7]. One of the alloys they studied, “2V Premendur”, is chemically similar to the modern Hiperco 50. They reported that the iron-nickel alloys optimized for very high initial permeability were the most sensitive to irradiation damage. The iron-cobalt alloy that they studied appeared to be unaffected. Gordon and Sery continued to study the iron-nickel alloys in various radiation fields and found that combinations of radiation and applied magnetic field could affect the magnetic domain alignment in much the same way as the combination of elevated temperature and magnetic fields [8, 9]. These studies suggest that radiation can influence microstructural-controlled magnetic properties analogously to thermal treatments even when the bulk temperature remains low. Research specific to modern Hiperco 50 has shown that operating above 450 °C, induces magnetic property degradations due to breakdown of the long-range order in the microstructure [10]. In his chapter on phase stability, Was [6] reviewed irradiation induced order-disorder reactions for several alloys and suggest that the influence of radiation on the ordering parameter was dependent on more factors than just fluence or dose rate and is therefore difficult to describe with one-dimensional radiation limits. Therefore a combination of elevated temperature and radiation environment would likely have synergistic impact on the phase stability in Hiperco 50 and the magnetic degradation reported by Lin [10] may occur at temperatures below 450 °C.

Hard Magnetic Materials and Radiation

The results of many permanent magnet radiation studies are summarized here. A more thorough review was presented in reference 11. Early radiation studies on SmCo magnets were performed using proton beam and proton induced neutron spallation radiation. Brown et al. found negligible deterioration in SmCo magnetic field after exposure to fluences up to 10^{18} n/cm² for neutrons with $E > 0.1$ MeV [12]. Yet nearly concurrent studies in France subjecting similar SmCo alloys to lower fluences ($\sim 10^{16}$ /cm² or 10^9 rad) of 400 GeV high energy protons resulted in nearly complete demagnetization [13]. Blackmore performed similar studies using fluences from 10^6 to 10^{10} rad of lower energy, 500 MeV, protons and distinguished the higher stability of the Sm₂Co₁₇ alloys compared to the earlier SmCo₅ alloys [14]. These results, shown in Figure 2(a), confirmed that radiation energy level was a key factor and

that both bulk chemistry (SmCo_5 versus $\text{Sm}_2\text{Co}_{17}$) as well as specific chemistry/processing (Crucore 18 versus other SmCo_5 magnets) were important.

Cost and Brown exposed NdFeB permanent magnet from different manufacturers while varying magnet geometry (and therefore demagnetization field), microstructural features, exposure temperature and radiation fluence [15, 16]. The experiments were conducted in fast neutron dominated spectrum, $E > 5$ eV, at temperatures of 77 and 153 °C, and at fluences up to nominally 6×10^{16} n/cm². Their results confirmed that irradiation induced degradation is sensitive to exposure temperature, chemistry, fabrication history, and the magnetic field shape/strength during irradiation. An important point made by the Cost and Brown reports was the need to specify experimental conditions when comparing results from various studies. They found remanence retention that ranged from 77% to 98% for different NdFeB magnets exposed to 4×10^{15} n/cm² for the same magnet shape and exposure conditions [16]. Figure 2(b) shows their NdFeB remanence results for samples exposed at 77 °C. Zeller studied magnetic field shape measured normal to the magnetization direction before and after elevated temperature and radiation exposure [17]. Temperature exposures up to 54 °C produces more severe demagnetization in the center of the specimen than near the edges. This same field-shape change was noted after exposure to ⁶⁰Co gamma radiation and to a more notable extent after irradiation with 106 MeV ²H charged particles. He noted that the presence of light elements such as boron should increase the susceptibility of the magnets to radiation-induced thermal spikes since the recoil energy is much higher for light elements.

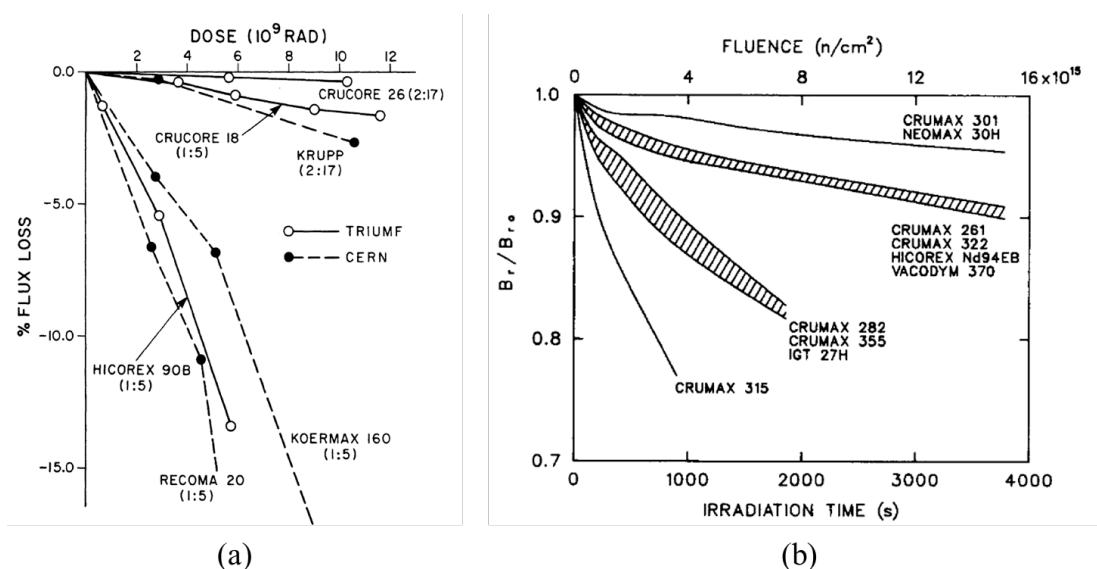


FIGURE 2. (a) Flux loss in SmCo_5 and $\text{Sm}_2\text{Co}_{17}$ magnets as a function of high energy proton fluence, compiled in reference 14. (b) Remanence loss as a function of neutron irradiation for various NdFeB alloys (b), from reference 16.

Finnish researchers delved further into the mechanisms of irradiation induced demagnetization in the 1990's [18, 19, 20]. This research began with positron defect measurements of NdFeB and NdFeB doped with Nb and Dy, irradiated from -258 °C to room temperature, and with magnetization along either the short or long sample dimension [18]. They found that the irradiation-induced sensitivity was greatly affected by temperature and not by defect formation. In a subsequent paper they proposed theoretical frame work for modeling the irradiation induced changes based on the energy of the impinging radiation particle being transferred to the lattice in a spherical region surrounding the primary knock-on atom [19]. The Kähkönen model suggests that the impinging radiation particle causes a local temperature above the Curie temperature and that the presence of a demagnetizing field can nucleate a reverse magnetic domain. In 1994 Kähkönen et al. published additional results that included magnetic packages rather than individual magnets, used impinging proton energy from 14 to 20 MeV, and studied the effects of alpha particle radiation at numerous temperatures from -251 to 222 °C [20]. Their theoretical work suggested that the correct combination of low demagnetization field and low bulk specimen temperature would preclude irradiation-induced demagnetization. They theorized and provided limited experimental verification that irradiation damage could be avoided by keeping the global temperature below 50 percent of the Curie Temperature for a particular

$\text{Nd}_2\text{Fe}_{14}\text{B}$ magnet with a fine grain structure and a low demagnetization field. Volk reviewed permanent magnet radiation studies, emphasized the theoretical framework presented by Kähkönen et al., and proposed research to test the Kähkönen theory [21]. He proposed radiation testing magnetic materials with the same grain size and known magnetization direction but with significantly different coercivity. Volk and colleagues emphasized testing magnetic materials in component configuration so that the demagnetization fields would be representative of components and proposed a model magnet configuration that was a variation on the offset-quadrupole magnet design [22]. Unfortunately this verification work has not been carried out.

Other radiation studies also support the Kähkönen proposed model. Alderman and Job irradiated NdFeB magnets with x-rays, gamma-rays, fast neutrons up to $1.6 \times 10^{14} \text{ n/cm}^2$, and with thermal neutrons up to $3.3 \times 10^{12} \text{ n/cm}^2$ [23, 24]. The fast neutron irradiation from 1-2 MeV ^{252}Ca exposure revealed no significant changes in flux densities after $1 \times 10^{12} \text{ n/cm}^2$ exposure, but there was a 0.6% and 10% flux density degradation respectively after the 2×10^{13} and $1.6 \times 10^{14} \text{ n/cm}^2$ exposure. Thermal neutron dominated irradiation was achieved with polyethylene moderation of the ^{252}Ca . It was found that thermal fluences of up to $3.34 \times 10^{12} \text{ n/cm}^2$ had no statically significant effect on residual induction. Chen et al. studied the effect of neutron irradiation on $\text{Sm}_2\text{Co}_{17}$ and NdFeB magnets using a water-moderated reactor with a thermal dominated spectrum [25]. For both NdFeB and SmCo-type magnets the measured bulk temperature was found to be dependent on the neutron flux. The recorded temperature in the NdFeB sample was 266°C for the neutron flux of $2.1 \times 10^{13} \text{ n/cm}^2/\text{s}$ and the NdFeB samples had no post-irradiation magnetic flux. The $\text{Sm}_2\text{Co}_{17}$ magnets, however, had imperceptible change in magnetic flux after up to $\sim 10^{20} \text{ n/cm}^2$. Figure 3 summarizes some of the irradiation experiments discussed, including the most radiation sensitive of the NdFeB alloys from [16]. The Curie temperatures of the Cost and Brown NdFeB samples were not stated and so the N38H T_c was used. There was measurable post-irradiation remanence degradation in each case for the samples were the bulk temperature during irradiation exceeded 50% of the T_c . Thus it is suggested that, in absence of targeted experiments to define precise temperature/radiation limits, the permanent magnets should not considered for use at a bulk temperature exceeding 50% of their Curie. If the demagnetization field of the magnetic component is large, or the flux rate is high, then a lower bulk temperature may be necessary.

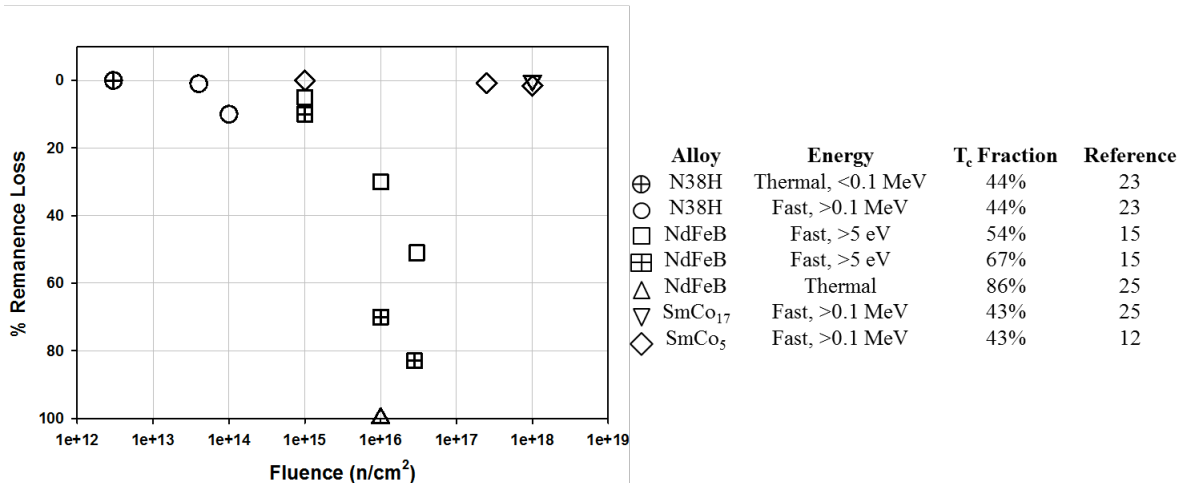


Figure 3. Summary of remanence loss as a function of neutron radiation fluence for rare earth permanent magnets. T_c fraction is bulk temperature reported during irradiation relative to Curie Temperature. The magnet shape, hence demagnetization fields, and the flux rates were not constant across these studies.

SUMMARY AND CONCLUSIONS

The research of radiation effects on magnetic materials was sparse before the 1980s and was focused on insertion devices for synchrotron radiation facilities. In recent decades, numerous researchers investigated rare earth magnets exposed to neutrons, electron beams, protons, gamma-radiation, and X-radiation. Studies have shown that accumulation of neutron or high energy particle radiation can degrade magnetic permanence. Numerous studies suggest that radiation damage affects both soft and hard magnetic properties in a manner that is analogous to and additive with thermal damage. Alloys with highly engineered microstructural features obtain the most desirable magnetic properties, but may be the most sensitive to thermal/radiological energy exposure.

Currently no verified models or experimental data are available to fully predict the effect of thermal and radiation conditions on magnetic materials in space power applications. More studies have addressed the radiation tolerance of hard magnetic materials than soft magnetic materials. There is a general literature consensus supporting the Kähkönen model that bulk temperature, radiation, and demagnetization fields all influence permanent magnet radiation hardness. A guideline of irradiation at bulk temperatures below 50% of Curie temperature in a specific NdFeB-type permanent magnet was suggested [20]. Note that this was a theoretical estimate with very specific assumptions including the size of reverse magnetic domains and the strength of the demagnetizing field. Several studies have pointed out that it is difficult to separate the intertwined chemical, microstructural, thermal, and radiological aspects of the radiation-induced loss of magnetic permanence [16, 20, 22]. Yet, as seen in Figure 3, there is experimental evidence that supports the notion of maintaining a low temperature fraction to avoid irradiation-induced demagnetization. Note that this 50% T_c temperature limit is suggested only as a guideline for initial design studies and is most appropriate for relatively low fluxes and low demagnetization fields. Specific magnetic material choices must be verified as radiation tolerant if designated for a specific mission.

Radiation damage in soft magnetic materials has not been studied as extensively as the damage in hard magnetic materials. The most extensive radiation review suggested that the more highly engineered alloys with the highest initial permeability were the most susceptible to radiation induced degradation [7]. However, the data was reported for fluences of 10^{18} n/cm² and higher. A reasonable supposition would be that the degradation mechanisms affecting soft magnetic materials would be dominated by the energy transfer. The published Curie temperature for candidate alloy Hiperco 50 is 938 °C. If the 50% T_c limit discussed relative to hard magnets could be applied, the combined conservative radiation-temperature limit would be ~ 330 °C. However Hiperco 50 experiences discontinuities in the B-H curve after very short times at 580 °C and induction loss after extended times at 450 °C [10]. In a review of phase transformations in radiation environments, the influence of thermal spike distribution on order-disorder transformation was described [6]. The fundamental mechanism appears to be the same as that driving reverse domain nucleation in the hard magnetic material, although the particular temperature/fluence relation need not be the same. In the absence of specific temperature/radiation interaction data, limiting Hiperco 50 application in a radiation environment to a bulk temperature of 160 °C (~50% of the general temperature limit) should be a conservative guideline to maintain optimum magnetic properties. Higher temperature applications are possible if the design can accommodate higher core losses resulting from decreased induction and increased coercivity. Studies on magnetic loss due to temperature effects alone can provide non-conservative guideline [10, 26] and additional performance losses can be anticipated due to radiation interactions for temperatures greater than 160 °C. The magnitude of the additional degradation could be estimated from modeling the radiation capture thermal distribution or could be measured experimentally.

NOMENCLATURE

B, **B** = magnetic field, magnetic field strength also known as induction strength or flux density
H, **H** = external magnetic field, external field strength also known as applied field strength
M, **M** = material magnetic field, material magnetic field strength
B_s, **M_s** = magnetic saturation strength
B_r, **M_r** = residual induction strength, remanence
H_c = coercivity, the reverse external magnetic field strength required to return induction strength to zero
H_{ci} = intrinsic coercivity, the reverse field strength required to return material magnetic strength to zero
μ_i, **μ_o** = initial permeability, permeability in vacuum
T_c = Curie temperature, temperature at which thermal energy overcomes magnetic domain alignment
n = neutron

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